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DISTRIBUTION OF RADIUM BETWEEN CRYSTALS OF LANTHANUM FLUORIDE AND THEIR SATURATED SOLUTION IN 3 PERCENT NITRIC ACID AT A TEMPERATURE OF 100°C

Acad V. G. Khlopin, M. S. Merkulova Radium Inst, Acad Sci USSR STAT

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Results of the type described in this paper can be applied in technical procedures whose purpose is concentration of radioactive elements. Tables referred to are appended.

The system studied by us previously -- isotopes of thorium and lanthanum fluoride (1) -- while similar to the yttrium-fluoride and calcium-fluoride system, is nevertheless distinguished to a certain extent from the latter because the cation valencies are not identical in the two systems. We were interested in studying either the YF3 - CaF2 system itself or a like system.

From the experimental viewpoint, an investigation of the YF_3 - CaF_2 system at present involves great difficulties; as a result we selected the completely similar LaF_3 - RaF_2 system, which is considerably more simple to investigate.

To study the distribution of radium between the crystals and solution of LaF3 in 3 percent HNO3, we prepared a suspension of LaF3 exactly as we described in our earlier work (1) and employed the same methods to conduct the experiments. Radium was determined in all cases by the emanation method on the apparatus developed by Ye. S. Shchepot'yeva (2) at the Radium Institute, Academy of Sciences USSR; we estimated the accuracy of these determinations to be ±1 percent.

The titre of the inactive suspension employed in the present work was equal to 1.3 mg Ia in one ml suspension. The lanthanum content in both inactive and active solutions was identical, being equal to 0.94 mg Ia in one mg solution.

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To study the distribution of radium, we used one or two drops of a titrated solution of a radium salt, added to the original solution of LaF3 in 3 percent HNO3. To prepare the titrated radium salt solution, we used a high-percent radium preparation in which the admixture of barium did not exceed 25 percent.

Since radium has no more stable isotopes, we employed an admixture to the radium consisting of continuously increasing quantities of its closest chemical analogue, barium, which is always truly isomorphic with it, so as to study the variation of concentration of the distributed substance within wide limits. Already in 1940, in the investigation of the reverse system of methylene blue +Ra(NO₃)₂, we showed that it was admissible and possible to make such a replacement of radium with barium for our purposes (3).

As usual, we first investigated the rate with which equilibrium was attained in the LaF3 - RaF2 system. The results are presented in Table 1.

The results from Table 1 show that equilibrium between LaF3 and the solution in relation to radium is established for practical purposes after 48 hr, while after 8 hr of recrystallization the value K4(D) can already be calculated as the average of the distribution data given in the second and third columns of Table 1.

For performing all further experiments, data of Table 1 permitted us to choose the time of recrystallization which would guarantee the attainment of equilibrium. None the less, we checked experimentally to see how well the values of Ks(D), obtained for the time we chose, would correspond with each other if we approached them from two opposite sides. As an example, we cite certain data (cf. Tables 2 and 3).

The results of Tables 2 and 3 show conclusively that the time of recrystallization which we chose (60 hr) completely guarantees that equilibrium will be attained with respect to radium between the solid crystalline phase and the solution and that the average values for Kq(D), obtained by reaching the equilibrium from two opposite sides, will coincide within the limits of experimental error.

To explain whether or not the distribution of radium between the crystals and solution of LaF3 takes place strictly according to the law for the distribution of a substance between two immiscible solvents, we studied within wide limits the effect on the distribution constant K4(D) of the variation of the concentration of the distributed substance. The results are shown in Table 4. These results show graphically that the value for the distribution constant K(D) of radium depends on the concentration of the distributed substance and decreases rapidly as the latter concentration is decreased. In this case, the value of $K_{\bullet}(\bar{D})$ clearly tends toward zero.

Thus, we now have the right to assert that Type III isomorphism, like isomorphism of Type II, is distinguished from Type I, according to the classification of V. Goldschmidt, principally by the fact that when substances which are mutually isomorphous, according to types III and II, form mixed crystals, a lower limit of miscibility is observed which is never observed for isomorphism of Type I. Consequently, when these mixed crystals are formed, whole elementary nuclei of the crystal lattice are replaced, as distinguished from replacement of ion for ion, atom for atom, or molecule for molecule which occurs in the case of Type I isomorphism.

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· /Tables follow.7

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Table 1. Attainment of Equilibrium for Distribution of Ra Between Crystals of LaF3 and Its Saturated Solution in 3% HNO3 When Temp =100°C

Time of Recrystal	K∮(D) Active Solution+ Inactive Suspension	K4(D) Inactive Solution + Active Suspension
1	4.6	51.0
4	7.8	41.3
8	10.0	30.4
24	15.6	22.7
48	17.4	19.4
72	18.4	18.0
120	18.2	18.6

Table 2. Distribution of Ra Between an Active Saturated Solution of LaF₃ in 3% HNO₃ and Inactive Crystals of LaF₃ When Temp = 100°C (total content of Ra + Ba: 3 mg in 30 ml solution)

			Coefficient	
% La in Solid Phase	% Ra in Solid Phase	Enrichment y/x	Impoverishment (100-y)/(100-x)	Distribution K(D)
9.31 18.37 27.56 27.56 32.13	65.24 79.96 86.30 87.80 89.96	7.01 4.35 3.13 3.18 2.80	0.38 0.25 0.19 0.17 0.15	18.2 17.4 16.5 18.7 18.7
		•	Average	18.1

Table 3. Distribution of Ra Between an Inactive Saturated Solution of LaF₃ in 3% $\rm HNO_3$ and Active Crystals of LaF₃ When Temp = $100^{\circ}\rm C$ (total content of Ra + Ba: 30 mg in 30 ml solution)

% La in Solid Phase	% Ra in Solid Phase	Enrichment y/x	Coefficient Impoverishment (100-y)/(100-x)	Distribution K(D)
7.80 15.63 23.15 35.00	60.0 78.19 83.96 91.02	7.7 5.0 3.63 2.60	0,43 0,26 0,20 0,14	17.8 19.2 18.1 18.6
			Averag	e 18.4

Table 4. Dependence of Average Distribution Coefficient K(D)
Between Crystals of IaF3 and Its Solutions in 3% HNO3
for Radium on Total Concentration of Ions of Ra··+ Ba··
in the Solution When Temp = 100°C

	Total Concentration	
Of Ions Ra Ba	Of Molecules RaF3 + BaF3 in mM	Value of K(D)
7 · 10°1 2.3 · 10°1 2.3 · 10°2 2.3 · 10°4 2.3 · 10°6 2.3 · 10°6	5.6 · 10 ⁻¹ 1.9 · 10 ⁻⁶	18.2 13.1 5.2 1.01 0.25 0.07
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